Signatures of the excitonic memory effects in four-wave mixing processes in cavity polaritons

Yu. P. Svirko and Makoto Kuwata-Gonokami*

Department of Applied Physics, Faculty of Engineering,

University of Tokyo and Cooperative Excitation Project of ERATO (JST),

7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan

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Abstract

We report the signatures of the exciton correlation effects with finite memory time in frequency domain degenerate four-wave mixing (DFWM) in semiconductor microcavity. By utilizing the polarization selection rules, we discriminate instantaneous, mean field interactions between excitons with the same spins, long-living correlation due to the formation of biexciton state by excitons with opposite spins, and short-memory correlation effects in the continuum of unbound two-exciton states. The DFWM spectra give us the relative contributions of these effects and the upper limit for the time of the exciton-exciton correlation in the unbound two-exciton continuum. The obtained results reveal the basis of the cavity polariton scattering model for the DFWM processes in high-Q GaAs microcavity.

Importance of the effects associated with the exciton-exciton interaction in the nonlinear optical response at the fundamental band edge has been revealed in a number of experiments in $\chi^{(3)}$-regime. These effects, which originate from the electromagnetic Coulomb interaction between photo-generated carriers and Pauli exclusion principle, are referred to as four particle correlation effects [1,2]. Extensive effort has been devoted to investigate such correlations in semiconductors by four-wave mixing spectroscopy, which gives us a unique opportunity to evaluate limits of applicability of the fermionic mean field theory [3]. However, under actual experimental conditions, the nonlinear optical measurements are affected by a number of intrinsic and extrinsic effects, which are caused by non-electronic degrees of freedom. These effects can not be discriminated in a nonlinear optical experiment in wide spectral region, preventing the direct comparison between theory and experiment.

However, the situation becomes more transparent when we restrict ourselves to the spectral window close to the exciton resonance, where the effects of four particle correlation can be classified in terms of spin dependent interaction between excitons [4–6]. In particular, a simple model [6], referred to as the weakly interacting boson (WIB) model, in which the interaction between $1s$ excitons is described by using two parameters to account for the interaction between excitons with the same and opposite spins, has allowed us to reproduce the overall features of the polarization-sensitive DFWM spectra.

The remarkable efficiency of the WIB model in the describing the polarization-sensitive DFWM measurements in time [7] and frequency domain [6] has made it a promising and handy scheme to study the correlation effects by methods of nonlinear spectroscopy. However, the underlying physics and, especially, the role of second,- and higher-order in exciton-exciton interactions effects [8], still remain unclear because the model does not account for memory effects. These effects are responsible for the temporal evolution of the 2ω-coherence within the continuum of the unbound two-exciton states and the long-living correlation due to the existence of biexciton, which has also been found to be important in FWM at semiconductor band edge [9–11]. Note, that the evolution equation for the excitonic polarization with account for the biexciton state has been obtained in [12] as a natural extension of the

WIB model. Similar semi-phenomenological treatment, which ignores the memory effects due to the unbound two-exciton continuum, can be developed by starting from the fermionic description of the excitonic nonlinearity [2,10,13]. The study of the effects of the bound and unbound two-exciton states in the third-order optical response is especially interesting in GaAs system, where - in strong contrast to the wide gap semiconductors such as CuCl [14] or ZnSe [15] - the biexciton binding energy is the order of the exciton linewidth [9].

In order to elucidate the role of the memory effects at the fundamental band edge, a special attention should be paid to the choice of the material for the nonlinear-optical measurements. Specifically, the experiment should be performed in a system where effects associated with the finite exciton population and inhomogeneity are minimized. In this condition the effects of the exciton population can be minimized by performing pump-probe experiment at large detuning from the excitonic resonance. In this condition the polarization-sensitive shift of the excitonic resonance, which is referred to as the optical Stark effect, is the signature of the four-particle correlation [16]. At resonance condition, the exciton-cavity coupled system is a well suited candidate to observe the signature of the four-particle correlation. This is because strong coherent coupling between exciton and photon in the high-Q microcavity, which leads to the formation of cavity polaritons [17] and suppresses incoherent effects in the DFWM signal [18]. In this paper, we show that the study of polarization-sensitive DFWM spectra in GaAs/AlGaAs QW embedded in the high-Q microcavity allows us to discriminate the memory effects. By comparing the results of the experiment and theory, we show that the correlation memory time of excitons in GaAs is very short justifying the description of the DFWM process, which is based on cavity polariton scattering. We also obtain the relative contributions from the bound and unbound two-exciton states to the nonlinear optical response.

The semiconductor microcavity investigated in this work has a single 12-nm-thick GaAs quantum well at the antinode of a $\lambda/2$-planar microcavity, consisting of 22 and 14.5 pairs of distributed Bragg reflectors for the bottom and topside respectively. All the measurements were performed at 13 K. The linear reflection spectrum shows that the normal mode splitting

at zero detuning (4.3 meV) is larger than the linewidth of either exciton or cavity mode (1.5 meV). The DFWM signal was measured in *(xxx)*-, *(xyy)*-, *(+++)*-, *(x++)*- and *(x+-)*-configurations, which are abbreviated by the polarizations of the pump, test and signal beams, respectively, in the self-pumped phase conjugation geometry [6]. We used the tunable picosecond pulses (pulse width of 1.9 psec and spectral width of 0.7 meV) from a Kerr-lens mode-locked Ti:Sapphire laser with a 76 MHz repetition rate. In order to ensure that the measurements were performed in the $\chi^{(3)}$-regime, we examined excitation-power dependence of the DFWM signal and found that it is proportional to $I^2_{pump}I_{test}$. The detailed description of the experiment can be find in [6].

The DFWM spectra at zero exciton-cavity detuning are presented in Fig.1a for different polarization configurations [6]. In the *(x++)*- and *(+++)*-configurations, the spectra experiment show nearly the same intensities for upper and lower polaritons. In the *(x+-)*-configuration, where the DFWM signal is dominated by the cavity polariton scattering is due to two-exciton states with zero angular momentum, the signal at lower mode is found to be 2.5 times of that at upper mode. The DFWM spectra in *(xxx)*- and *(xyy)*-configurations show a switching between upper and lower modes.

In order to explain these results, we need to examine how the memory effects in the exciton interaction manifest themselves in different polarization configuration. Following [11] we consider the resonant excitation only and start from the equation of motion for the normalized complex amplitudes of the right- and left-circular components of the excitonic polarization at the frequency ω, $p_\pm =< b_\pm > /(Vv_e)^{1/2}$, where $b_\pm$ is the exciton annihilation operator, V and v_e are the crystal and exciton volume respectively, subscripts " $\pm$ " label right- and left-circular components. The exciton volume is defined from the conventional relationship between the exciton and interband dipole moments: $\mu_{ex}/\mu = (V/v_e)^{1/2}$. The evolution equation for $p_\pm$ can be presented in the following form:

$$-i\frac{\partial p_\pm}{\partial t} + \Delta p_\pm = (1 - C|p_\pm|^2)\Omega_\pm + \\ -p^*_\pm \int_0^\infty F(\tau)p^2_\pm(t-\tau)e^{-2i\Delta\tau}d\tau - p^*_\mp \int_0^\infty G(\tau)p_+(t-\tau)p_-(t-\tau)e^{-2i\Delta\tau}d\tau \quad (1)$$

Here $\Omega_\pm = \mu E_\pm/\hbar$ are the Rabi frequencies, which correspond to the right- and left-circular components of the electric field $E_\pm$ of the light wave at the QW, $\Delta = \omega_e - \omega - i\gamma$, ω_e and γ are the exciton frequency and dephasing rate, respectively; $C > 0$ is the phase space filling (PSF) constant [19]; $F(\tau)$ and $G(\tau)$ are memory functions, which account for both instantaneous and retarded parts of the exciton-exciton interaction.

Since in our experiment, the pulse is longer than the exciton dephasing time, we can consider the steady state approximation ignoring the time dependence of slowly varying amplitudes $p_\pm$ and $\Omega_\pm$ in Eq. (1). By using Eq. (1) and the evolution equation for the electric field at the QW [12,22], the steady-state amplitudes of the third-order polarization at the frequency ω, which is responsible for the DFWM signal in the phase conjugated geometry, can be presented in the following form:

$$p^{(3)}_\pm = A\{E^2_{\pm,pump}[C\Delta + \int_0^\infty F(\tau)e^{-2i\Delta\tau}d\tau] + E^2_{\mp,pump}\int_0^\infty G(\tau)e^{-2i\Delta\tau}d\tau\}E^*_{\pm,test} \qquad (2)$$

where A accounts for resonance enhancement of the electric field in the microcavity, $E_{\pm,pump}$ and $E_{\pm,test}$ are amplitudes of the electric field associated with the pump and test beams, respectively, at the QW.

In order to show how the memory effects manifest themselves in the nonlinear response, we separate the memory function $F(\tau)$ in terms of instantaneous (mean field) part given by $\phi > 0$, and retarded (correlation) part [11], which is given by $\Phi(\tau)$: $F(\tau) = \phi\delta(\tau) - \Phi(\tau)$. The mean field parameter ϕ is of the first order in exciton-exciton interaction and describes the interaction between excitons with same spins and zero center-of-mass momentum [20,21], while $\Phi(\tau)$ accounts for correlation effects of the second- and higher-order of the interaction between two excitons. The memory function $G(\tau)$ accounts for the effects arising from the interaction between excitons with opposite spins. In this case, the first-order in exciton-exciton interaction term vanishes and this memory function contains correlation effects of the second- and higher order in interaction between two excitons. In this configuration, there exists a bound state of two excitons. Correspondingly, we separate $G(\tau)$ in terms of contribution from the unbound two-exciton part given by $\Psi(\tau)$ and biexciton part given by

$\psi e^{i\omega_B\tau}$, where ω_B is the biexciton binding energy: $G(\tau) = \Psi(\tau) - i\psi e^{i\omega_B\tau}$ [11].

In the high-Q microcavity, the strong coupling between excitons and photons produces polariton modes, which dominate both linear and DFWM spectra. These modes are referred to as lower and upper cavity polaritons. At zero exciton-cavity detuning their frequencies are $\omega_{\alpha,\beta} = \omega_e \mp g$, respectively, where $g = (2\pi\omega\mu^2/\hbar n v_e)^{1/2}$ is the energy of the dipole coupling between exciton and photon and n is the refractive index. The intensities of the polarization-sensitive DFWM signal at frequencies of the lower and upper polaritons for different polarization configurations can be obtained from (2) as follows: $I^{xxx}_{\alpha,\beta} \propto |R + W \pm (Cg + \delta R + \delta W)|^2$, $I^{xyy}_{\alpha,\beta} \propto |R - W \pm (Cg + \delta R - \delta W)|^2$, $I^{+++}_{\alpha,\beta} \propto |R \pm (Cg + \delta R)|^2$ and $I^{x+-}_{\alpha,\beta} \propto |W \pm \delta W|^2$, where

$$\begin{aligned}
R &= \phi - \int_0^\infty \Phi(\tau)e^{-2\gamma\tau}\cos 2g\tau d\tau \\
\delta R &= i\int_0^\infty \Phi(\tau)e^{-2\gamma\tau}\sin 2g\tau d\tau \\
W &= \int_0^\infty \Psi(\tau)e^{-2\gamma\tau}\cos 2g\tau d\tau + \frac{(2ig + \omega_B)\psi}{(2i\gamma + \omega_B)^2 - 4g^2} \\
\delta W &= i\int_0^\infty \Psi(\tau)e^{-2\gamma\tau}\sin 2g\tau d\tau + \frac{2g\psi}{(2i\gamma + \omega_B)^2 - 4g^2}
\end{aligned} \tag{3}$$

Normal mode splitting $2g$ is the major spectral characteristic of the strongly coupled exciton-cavity system and, correspondingly, the role of the memory effects in the excitonic nonlinear response is determined by its ratio to the spectrum width of the memory functions. In order to clarify the role of the memory effects in the DFWM spectra, we first examine the long-time memory limit case, i.e. $2g >> \tau_c^{-1}$, where τ_c is the correlation time of the memory functions $\Phi(\tau)$ and $\Psi(\tau)$. By simplifying Eq. (3) with account for $2g\tau_c >> 1$ and $g >> \gamma, \omega_B$ one can arrive at the following equations for the polariton intensities: $I^{+++}_{\alpha,\beta} \propto |\phi \pm Cg|^2$, $I^{xxx}_{\alpha,\beta} \propto |\phi \pm (Cg - \psi/2g)|^2$ and $I^{xyy}_{\alpha,\beta} \propto |\phi \pm (Cg + \psi/2g)|^2$. Since $\phi, \psi > 0$ and $\gamma, \omega_B < 2g$ one may see that $I^{xyy}_\alpha > I^{+++}_\alpha$ (intensity of the lower polariton in *(xyy)*-configuration is higher than that in *(+ + +)*-configuration) and $I^{xxx}_\beta > I^{+++}_\beta$ (intensity of the upper polariton in *(xxx)*-configuration is higher than that in *(+ + +)*-configuration). However, it can be clearly observed from the spectra 1n Fig. 1a, that such a conclusion contradicts to the experimental

results making assumption $2g\tau_c >> 1$ is invalid.

Therefore, our experimental findings invoke the condition $2g < \tau_c^{-1}$. In such a case, we can substitute $e^{-2\gamma\tau}cos2g\tau \approx 1 - 2\gamma\tau - 2g^2\tau^2 + ...$ and $e^{-2\gamma\tau}sin2g\tau \approx 2g\tau + ...$, and neglect δR in comparison with Cg. Similarly, at $\omega_B < 2g$ one can estimate $W \approx \int_0^\infty \Psi(\tau)d\tau$ and $\delta W \approx -\psi/2g$. These gives $I^{xxx}_{\alpha,\beta} \propto |R + W \pm (Cg + \delta W)|^2$, $I^{xyy}_{\alpha,\beta} \propto |R - W \pm (Cg - \delta W)|^2$, $I^{+++}_{\alpha,\beta} \propto |R \pm Cg|^2$ and $I^{x+-}_{\alpha,\beta} \propto |W \pm \delta W|^2$ ensuring $I^{+++}_{\alpha} > I^{xyy}_{\alpha} > I^{xxx}_{\alpha}$ and $I^{+++}_{\beta} > I^{xxx}_{\beta} > I^{xyy}_{\beta}$. Note that since we observe $I^{x+-}_{\alpha} > I^{x+-}_{\beta}$, the following relationship holds: $W < \delta W < 0$. The observed difference in the intensities of the upper and lower polaritons in the *(x+-)*-configuration originates from the bound two-exciton state. The ratio $I^{x+-}_{\alpha}/I^{x+-}_{\beta} \approx 2.5$ obtained in the experiment is consistent with the followed from the sum rule [11] theoretical estimation $I^{x+-}_{\alpha}/I^{x+-}_{\beta} \approx 1 + 2\omega_B/g$, for typical GaAs biexciton binding energy [9]. The calculated DFWM spectra with account for the bound and unbound two-exciton states are presented in Fig. 1b for $(-W) : \delta W : R : Cg = 0.7 : 0.23 : 0.12 : 1$. We would like to note here, that our estimation $\tau_c < (2g)^{-1}$ is consistent with the results of the calculation of the memory functions within the 1D-Hubbard model framework [11].

With account for $\delta R << Cg$ and $|\delta W| < |W|$, the obtained result returns the prediction of the polariton scattering model [6], which has allowed us obtain Eq. (2) with the frequency independent W and R. In this model, these parameters account for the attraction and repulsion between excitons with opposite and same spins, respectively, in the phenomenological WIB Hamiltonian [6,23]. The experimental spectra at both zero (see Fig. 1a) and arbitrary detuning for the above mentioned polarization configurations have been explained by the following relationship between the parameters of the polariton scattering model: $(-W) : R : Cg = 0.75 : 0.1 : 1$ [6]. This has allowed us to conclude that the attractive interaction between excitons and the PSF effect dominate in the DFWM process in the high-Q microcavity. Apparently the polariton scattering model failed to explain the difference in I^{x+-}_{α} and I^{x+-}_{β}, which is due to the biexciton effect. Nevertheless, the parameters, which has been obtained in [6], coincide with our present estimations, because the biexcitonic effects do not affect significantly the spectra in *(xxx)*- and *(xyy)*-configurations.

The relatively small value of the parameter R obtained in the experiment is due to nearly cancellation of the first- and higher-order in the exciton-exciton interaction contributions [8] to the resonant third-order susceptibility. This cancellation has also been discussed in [11] in terms of constraints, which are imposed on the spectral density of the memory functions by the sum rules.

In conclusion, we formulate the resonant DFWM results in terms of exciton memory functions and show that the polarization-sensitive DFWM spectra give us an important information on the memory effects in exciton-exciton interaction. The intensity of the DFWM signal in the strongly coupled exciton-cavity system is determined by both short-memory correlation in the unbound two-exciton continuum and long-memory correlation associated with the biexciton state. Both these effects give the second- and higher-order in exciton-exciton interaction contributions to the resonance optical nonlinearity, while the mean field, instantaneous contribution, which is of the first-order in the exciton-exciton interaction, is nearly canceled. By comparing the results of the experiment and theory in various polarization configurations, we estimate the upper limit of the correlation time of the memory functions, $\tau_c << 900$ fs, which describe the 2ω-coherence due to the continuum of the unbound two-exciton states. This also allows us to show that the relative contribution to the resonant third-order susceptibility from the biexciton in GaAs is about 30 percent. We show that the short memory time of the exciton-exciton interaction permits to describe the coherent optical response of the excitons in the high-Q semiconductor microcavity in terms of the cavity polariton scattering model, which should be extended to account for the biexciton state.

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REFERENCES

* To whom communication should be addressed. Email address: gonokami@ap4.t.u-tokyo.ac.jp.

[1] H. Wang, K. Ferrio, D. G. Steel Y. Z. Hu, R. Binder, and S. W. Koch, Phys. Rev. Lett. **71**, 1261 (1993); T. Saiki, M. Kuwata-Gonokami, T. Matsusue, and H. Sakaki, Phys. Rev. B **49**, 7818 (1994); P. Kner, S. Bar-Ad, M. V. Marquezini, D. S. Chemla, and W. Schaefer, Phys. Rev. Let. **78**, 1319 (1997).

[2] D. S.Chemla, in *Nonlinear Optics in Semiconductors I*, edited by R. K. Willardson, E. R. Weber, E. Garmiere, and A. Kost, Semiconductors and Semimetals Vol. 58 (Academic Press, San Diego, 1999).

[3] H. Haug and S. W. Koch. *Quantum Theory of the optical and Electronic Properties of Semiconductors*, (World Scientific, Singapore, 1993).

[4] V. Chernyak and S. Mukamel, J. Opt. Soc. Am. B **13**, 1302 (1996).

[5] M. Z. Maialle, E. A. de Andrada e Silva, and L. J. Sham, Phys. Rev. B **47**, 15776 (1993).

[6] M. Kuwata-Gonokami, Shin Inouye, H. Suzuura, M. Shirane, R. Shimano T. Someya and H. Sakaki, Phys. Rev. Lett. **79**, 1341 (1997)

[7] T. Aoki, G. Mohs, M. Kuwata-Gonokami, and A. A. Yamanishi, Phys. Rev. Lett. **82**, 3108 (1999).

[8] J. Inoue, T. Brandes and A. Shimizu. J. Phys. Soc. Jpn. **67**, 3384 (1998).

[9] D.J. Lovering, R. T. Phillips, G. J. Denton, G. W. Smith, Phys. Rev. Lett. **68**, 1880 (1992) ; E. J. Mayer, G. O. Smith, V. Heuckeroth, J. Kuhl, K. Bott, A. Schulze, T. Meier, D. Bennhardt, S.W. Koch, P. Thomas, R. Hey and K. Ploog, Phys. Rev. B **50**, 14730 (1994) ; B. F. Feuerbacher, J. Kuhl and K. Ploog. Phys. Rev. B **43**, 2439 (1991) ; X. Fan, H. Wang, H. Q. Hou, and B. E. Hammons, Phys. Rev. B **57**, 9451 (1998).

[10] W. Schaefer, D. S. Kim, J. Shah, T. C. Damen, J. E. Cunningham, K. W. Goossen, L. N. Pfeiffer, K. Kohler, Phys. Rev. B **53**, 16429 (1996).

[11] Th. Ostrreich, K. Schonhammer and L. J. Sham, Phys. Rev. Lett. **74**, 4698 (1995) ; Phys. Rev. B, **58**, 12920 (1998) ; L. J. Sham and Th. Ostreich, Journ. Luminescence (to be published).

[12] M. Kuwata-Gonokami, T. Aoki, C. Ramkumar, R. Shimano and Yu. P. Svirko, Journ. Luminescence (in press).

[13] P. Kner, W. Schaefer, R. Lovenich, and D. S. Chemla, Phys. Rev. Lett. **81**, 5386 (1998).

[14] G. M. Gale and A. Mysyrovicz, Phys. Rev. Lett. **32**, 727 (1974).

[15] M. Itoh, Y. Nozue, T. Itoh, M. Ueta, S. Satoh, and K. Igaki, Journ. Luminescence **18/19**, 568 (1979); Y. Nozue, M. Itoh and K. Cho, J. Phys. Soc. Jpn. **50**, 889 (1981); Q. Fu, D. Lee, A. Mysyrowicz, A. V. Nurmikko, R. L. Gunshor, and L. A. Kolodziejski, Phys. Rev. B **37**, 8791 (1988); T. Saiki, M. Kuwata-Gonokami, K. Ohkawa, and T. Mitsuyu, Solid St. Commun. **95**, 679 (1995).

[16] R. Shimano, M. Kuwata-Gonokami, Y. Kadoya, and M. Yamanishi, *Proceedings of the Quantum Electronics and Laser Science Conference (QELS'97)* (Optical Society of America, Baltimore, USA, 1997), p.199; C. Sieh, T. Meier, A. Knorr, S. W. Koch, P. Brick, M. Hubner, C. Ell, J. Prineas, G. Khitrova, and H. Gibbs, Phys. Rev. Lett. **82**, 3112 (1999).

[17] C. Weisbuch, M. Nibshioka, A. Ishikawa, and Y. Arakawa, Phys. Rev. Lett. **69**, 3314 (1992).

[18] M. Shirane, C. Ramkumar, Yu. P. Svirko, H. Suzuura, Shin Inouye, R. Shimano, T. Someya, H. Sakaki, and M. Kuwata-Gonokami, Phys. Rev. B **58**, 7978 (1998)

[19] O. Akimoto and E. Hanamura, J. Phys. Soc. Jpn. **33**, 1537 (1972).

[20] A. L. Ivanov, H. Haugh and L.V. Keldysh, Phys. Rep. **296**, 239 (1998).

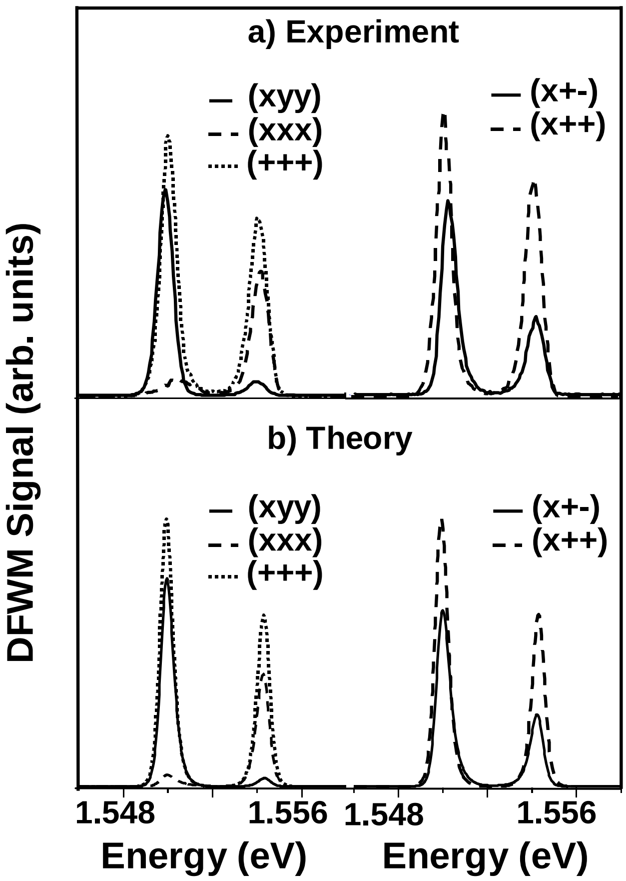
[21] E. Hanamura, J. Phys. Soc. Jpn. **37**, 1553 (1974).

[22] L. A. Lugiato, Progr. Opt. **21**, 71 (1984).

[23] H. Suzuura, Yu. P. Svirko, and M. Kuwata-Gonokami, Solid St. Commun. **108**,289 (1998) ; Yu. P. Svirko, M. Shirane, H. Suzuura, and M. Kuwata-Gonokami, J. Phys. Soc. Jpn. **68**, 674 (1998).

FIGURES

FIG. 1. Polarization-sensitive DFWM spectra. Left spectra: DFWM signal intensity *(xyy)*-, *(xxx)* and *(+++)*-configurations shown by solid, dashed and dotted lines, respectively. Right spectra: in *(x+-)*-and *(x++)*-configurations, shown by solid and dashed lines, respectively, and scaled by factor 4. (a) experiment; (b) calculated with account for the bound biexciton: $W : \delta W : R : g\nu = .7 : .23 : .12 : 1$.



a) Experiment
(xyy)
(xxx)
(+++)
(x+-)
(x++)
b) Theory
(xyy)
(xxx)
(+++)
(x+-)
(x++)
DFWM Signal (arb. units)
1.548
1.556
1.548
1.556
Energy (eV)
Energy (eV)